

Orbital Degeneracy and the Microscopic Model of the FeAs Plane in the Iron-Based Superconductors

Tao Li

Department of Physics, Renmin University of China, Beijing, 100872, P.R.China

(Dated: April 3, 2008)

A microscopic model for the FeAs plane in the newly discovered Iron-based superconductors is proposed with the emphasis on the role of the orbital degeneracy between the Fe $3d_{xz}$ and $3d_{yz}$ orbital in the crystal field environment. The model predicts commensurate antiferromagnetic ordered ground state for the parent compounds and a d-wave superconducting ground state at finite doping. Inter-orbital couplings plays an interesting role in enhancing both orders. Correlated variational wave functions are proposed for both ordered phases.

PACS numbers:

The newly discovered Iron-based superconductors F-doped LaOFeAs[1] and its derivatives[2, 3, 4, 5] with rare earth substitution arose many interests in the community. Similar to the high- T_c cuprates, the Iron-based superconductors also have a layered structure in which the role of the Copper oxygen plane in the high- T_c cuprates is played by the FeAs plane. In both systems, the charge dopant and the doped carrier are separated spatially which produces an ideal environment for the coherent motion of the latter at an incommensurate band filling. This similarity may imply that a still higher T_c is possible in this series of compounds by optimize the interlayer distance and planeness of the FeAs layer.

Band structure as calculated from Density functional theory(DFT) and its DMFT improvement had been examined by several groups[6, 7, 8, 9, 10]. Not to one's surprise, most of the spectral weight close to the Fermi energy are contributed by the Fe 3d orbital, as the Fe is the only ion in the system that posses a unclosed shell. The DFT calculation also predicted a antiferromagnetic ordered ground state for the parent compound LaOFeAs with a ordered moment about 2.3 Bohr magneton, which may also be consistent with some preliminary experiment results on this system.

Based on the results of the DFT calculations, several phenomenological theories for the doped Iron-based superconductors were proposed[11, 12, 13]. In this paper, we follow instead the quantum chemistry considerations and propose a tight binding microscopic model for the FeAs plane. In our model, the orbital degeneracy between the Fe $3d_{xz}$ and $3d_{yz}$ orbital plays an essential role. Our model predicts commensurate antiferromagnetic ordered ground state for the parent compound and a d-wave superconducting ground state at finite doping. Inter-orbital couplings plays an interesting role in enhancing both kind of orders. We also propose correlated variational wave functions for both phases.

A key issue for the construction of a microscopic model for the FeAs plane is to elucidate the role of the orbital degeneracy of the Fe 3d orbital. In the presence of crystal field, the five fold degenerate Fe 3d atomic orbital will

split according to the irreducible representations of the crystal symmetry and not all five 3d orbital are equally important for the low energy physics. According to the DFT calculation, the crystal splitting of the Fe 3d orbital is small and all five Fe 3d orbital should be included in the model in principle. However, the inclusion of the electron correlation effect can enhance the crystal splitting and vice versa. Here we assume the crystal field splitting to be sufficiently large for the discussion of the low energy physics and base our discussion solely on symmetry considerations.

In the FeAs plane, each Fe ion sits at the center of inversion of a squashed(along the normal of the FeAs plane) tetrahedron formed by four neighboring As ions(see figure 1). Using the lattice constants reported in the literature[7], one estimate that the Fe-As bond make a angle of about 58.8 degree with the normal of the FeAs plane, while in a perfect tetrahedron the corresponding angle should be 54.7 degree. In such a local environment of S_4 symmetry[9], the five fold degenerate 3d orbital split into three nondegenerate($3d_{3z^2-r^2}$, $3d_{x^2-y^2}$, and $3d_{xy}$) orbital and one doubly degenerate orbital($3d_{xz}$ and $3d_{yz}$)(see figure 2). If we assume that the crystal field is contributed mainly by the four neighboring As ions, one would expect the $3d_{3z^2-r^2}$ orbital to have the lowest energy, since the As tetrahedron in squashed in z-direction and the electron cloud of the $3d_{3z^2-r^2}$ orbital has the best chance to avoid the As ion(see figure 1). The next lowest energy orbital would be $3d_{xy}$ whose lobe points toward the vacancy between the As ions in the plane. If the tetrahedron is not squashed, the $3d_{xy}$ orbital would be degenerate with the $3d_{3z^2-r^2}$ orbital. The squashing distortion of the tetrahedron also lift the degeneracy between the remaining three orbital, namely $3d_{x^2-y^2}$, $3d_{xz}$, $3d_{yz}$. Since the tetrahedron around the Fe ion is squashed toward the plane, the $3d_{x^2-y^2}$ orbital should be higher in energy than $3d_{xz}$ and $3d_{yz}$ orbital. In the tetragonal structure of the FeAs plane, the degeneracy between the $3d_{xz}$ and $3d_{yz}$ orbital is protected by symmetry.

The divalent Fe ions in the FeAs plane have six elec-

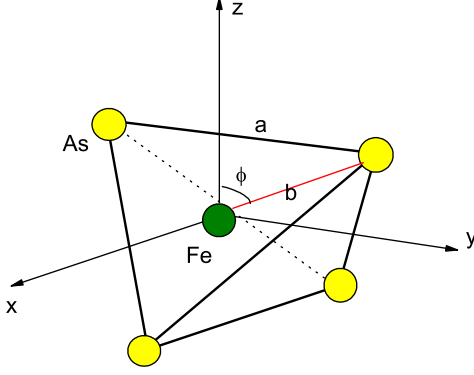


FIG. 1: The squashed tetrahedron formed by four neighboring As ion around each Fe ion in the FeAs plane. From the lattice constant reported in the literature, $a=4.020$ Å, $b=2.35$ Å, the angle ϕ between the Fe-As bond with the normal of the Fe-As plane is estimated to be 58.8 degree, while in a perfect tetrahedron the angle would be 54.7 degree. Here we use a coordinate system in which the x and y axis point along the As-As bond directions.

trons in their 3d shell. According to the above splitting scheme, one would expect a very peculiar situation to occur in the parent compounds in which the Fermi energy lies within two degenerate bands which are both half filled. We think this is the reason that the parent compounds differ from usual band insulator or band metal. It may also hold the key for the lightly doped LnOFeAs system to show such remarkable properties as having a T_c as high as 50 K[5].

In the following, we assume the crystal splitting to be large enough to neglect the filled $3d_{3z^2-r^2}$ and $3d_{xy}$ band and the empty $3d_{x^2-y^2}$ band in the discussion of low energy physics. With this simplification, we are left with a model with two degenerate bands. In this model, each Fe site can accommodate at most four electrons in the two degenerate bands. In the parent compound, each Fe site have two electrons on average in the two degenerate bands and the system is thus half filled. The interactions between the electrons on the same Fe site can be classified into three types, namely the intra-orbital Hubbard repulsion U , the spin independent part of the inter-orbital Hubbard repulsion U_1 , and the spin dependent part of the inter-orbital Hubbard repulsion J (the usual Hund's rule coupling). From the definition of these terms, one easily see that that the inequality $U > U_1 > \frac{J}{4} > 0$ should be satisfied. These interaction terms is represented by the model Hamiltonian

$$H_U = U \sum_i (n_{i,xz,\uparrow} n_{i,xz,\downarrow} + n_{i,yz,\uparrow} n_{i,yz,\downarrow})$$

$$+ U_1 \sum_i (n_{i,xz,\uparrow} + n_{i,xz,\downarrow})(n_{i,yz,\uparrow} + n_{i,yz,\downarrow}) - J \sum_i \vec{S}_{i,xz} \cdot \vec{S}_{i,yz} \quad (1)$$

in which $n_{i,xz,\uparrow} = c_{i,xz,\uparrow}^\dagger c_{i,xz,\uparrow}$ denotes the number of up spin electron in the $3d_{xz}$ orbital. $\vec{S}_{i,xz} = \frac{1}{2} c_{i,xz,\alpha}^\dagger \vec{\sigma}_{\alpha,\beta} c_{i,xz,\beta}$ denotes the spin density on the $3d_{xz}$ orbital. The meaning of other terms in the equation is similar.

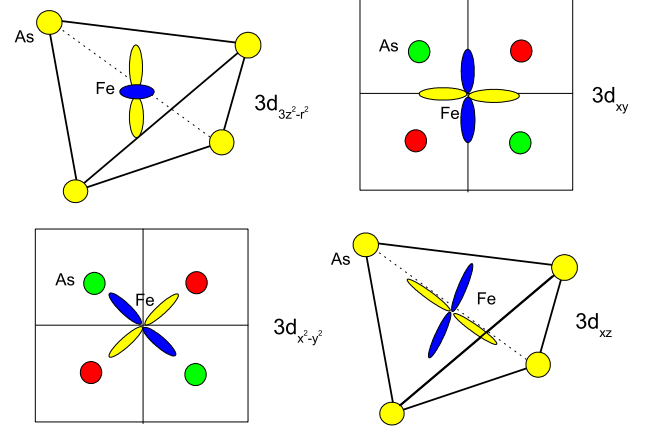


FIG. 2: The five d-orbital in the crystal field of the tetrahedron formed by the four neighboring As ions around each Fe ion. The $3d_{x^2-y^2}$ and the $3d_{xy}$ orbital are shown here in a projection onto the xy plane. In the projection graph, red(green) filled circles denote As ion above(below) the xy plane. Here we only show the $3d_{xz}$ orbital, the $3d_{yz}$ orbital can be obtained from the $3d_{xz}$ orbital through a rotation of $\frac{\pi}{2}$ along the z -axis.

In the FeAs plane, the Fe ions are too remote from each other to have direct hopping and the As ion is needed to bridge the hopping between the Fe ions. The As ion has three 3p orbital in its outmost shell. Since the $3p_z$ orbital is far away from the Fermi energy, we consider only the hopping path mediated by the $3p_x$ and $3p_y$ orbital of As ion. Symmetry considerations shows that the hopping mediated by the As $3p_x$ and $3p_y$ orbital conserve the orbital index, namely, an electron initially on the Fe $3d_{xz}$ ($3d_{yz}$) orbital can only hop onto the $3d_{xz}$ ($3d_{yz}$) orbital of destination Fe ion (the inclusion of the hopping path mediated by $3p_z$ orbital will break such a symmetry) (see figure 3). Another peculiarity about the As ion aided effective hopping is that the hopping integral between next-nearest-neighboring Fe ions is anisotropic (the hopping between nearest neighboring Fe ions remains isotropic). This anisotropy depends on the orbital and is a reflection of the anisotropic nature of the $3d_{xz}$ and $3d_{yz}$ orbital. Taking all these considerations into account, we arrive at the following model for the effective hopping between the Fe

ions.

$$\begin{aligned}
H_t = & -t \sum_{i, \vec{\delta}, \sigma} \left[(c_{i, xz, \sigma}^\dagger c_{i+\vec{\delta}, xz, \sigma} + c_{i, yz, \sigma}^\dagger c_{i+\vec{\delta}, yz, \sigma}) + H.C. \right] \\
& -t_1 \sum_{i, \sigma} \left[(c_{i, xz, \sigma}^\dagger c_{i+\vec{\delta}', xz, \sigma} + c_{i, yz, \sigma}^\dagger c_{i+\vec{\delta}', yz, \sigma}) + H.C. \right] \\
& -t_2 \sum_{i, \sigma} \left[(c_{i, xz, \sigma}^\dagger c_{i+\vec{\delta}'', xz, \sigma} + c_{i, yz, \sigma}^\dagger c_{i+\vec{\delta}'', yz, \sigma}) + H.C. \right], \quad (2)
\end{aligned}$$

in which $\vec{\delta} = \vec{e}_x$ or \vec{e}_y denotes the nearest neighboring vectors on the square lattice form by Fe ions. $\vec{\delta}' = \vec{e}_x + \vec{e}_y$ and $\vec{\delta}'' = \vec{e}_x - \vec{e}_y$ denotes the next nearest neighboring vectors. Since there are two hopping paths between the nearest neighboring Fe ions but only one for the next-nearest neighboring Fe ions, t is roughly a factor of two larger than both t_1 and t_2 . From symmetry considerations, one can also show that all of the three hopping integrals, t , t_1 and t_2 can be taken positive with a suitable choice of gauge. The dispersion relation of the $3d_{xz}$ band is given by

$$\begin{aligned}
E_{xz, k} = & -2t(\cos(k_x) + \cos(k_y)) - 2t_1 \cos(k_x + k_y) \\
& - 2t_2 \cos(k_x - k_y), \quad (3)
\end{aligned}$$

the dispersion relation of the $3d_{yz}$ band can be obtained from that of $3d_{xz}$ band by the reflection in momentum space $k_x \rightarrow -k_x$.

The total Hamiltonian of the system is the sum of the on-site term H_U and the hopping term H_t . Before looking at the phase diagram of the model, we first estimate the model parameters. According to DFT calculations, the Hubbard repulsion U and U_1 are about 4eV in magnitude and the Hund's rule coupling J is about 1eV in magnitude. For the hopping terms, as both the $\text{Fe}3d_{xz} - \text{As}3p_x$ separation and the hopping integral between the two are of the order of 1eV, we estimate the effective hopping integral between the Fe ions mediated by the As ions to be also of the order of 1eV. Thus the system has a moderate level of electron correlation and moderate level of frustration (as indicated by the ratio between t and t_1).

Now we discuss the possible phase diagram of the model. In the half filled parent compounds, each site is occupied on average by two electrons. Since the intra-orbital Hubbard repulsion U is larger than the inter-orbital Hubbard repulsion U_1 , the two electrons tend to occupy different orbital and the remaining Hund's rule coupling tends to align up the spin of the electrons on both orbital. Thus each individual Fe ion carries approximately a unit spin angular momentum and shows two Bohr magneton of magnetic moment. However, the inclusion of the hopping terms between the Fe ions will induce antiferromagnetic spin exchange between neighboring Fe ions within each band. Since the frustration is moderate, one can expect an commensurate antiferromagnetic order to appear in the ground state of parent

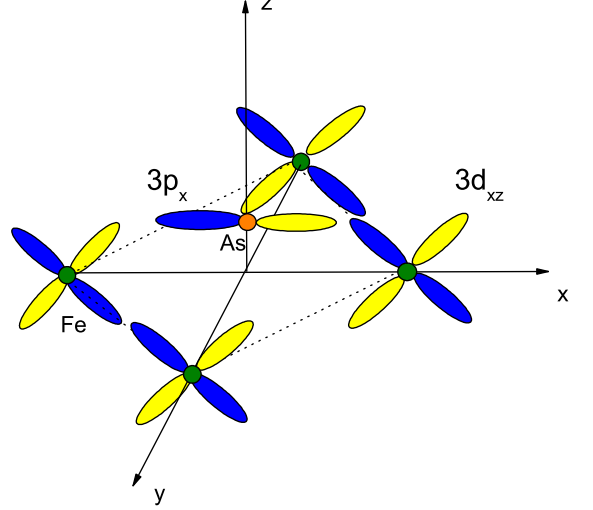


FIG. 3: A schematic representation of the As 3p orbital aided hopping between neighboring Fe ions on the square lattice. Note that the As ions lies above or below the Fe plane. Simple symmetry consideration shows that the electron on Fe $3d_{xz}$ ($3d_{yz}$) orbital can hop only through the As $3p_x$ ($3p_y$) orbital. This explains the conservation of orbital index and the anisotropy of the hopping Hamiltonian. The electron wave function on the yellow (blue) lobes have positive (negative) values.

compound. As a result of the Hund's rule coupling, the antiferromagnetic order in the $3d_{xz}$ and the $3d_{yz}$ band enhance with each other. With this understanding in mind, we can write down the following variational wave function for the ground state of the half filled system,

$$|\Psi\rangle = |\text{SDW}\rangle_{xz} \otimes |\text{SDW}\rangle_{yz}, \quad (4)$$

in which $|\text{SDW}\rangle_{xz}$ and $|\text{SDW}\rangle_{yz}$ denote the SDW ordered state in the $3d_{xz}$ and the $3d_{yz}$ band.

In our model, the two bands are correlated with each other through the inter-orbital Hubbard term and the Hund's rule coupling. To describe such correlation effect, we introduce the following Jastrow-type variational wave function

$$|\Psi\rangle = g^{\hat{D}} g_1^{\hat{V}} g_J^{\hat{S}} |\text{SDW}\rangle_{xz} \otimes |\text{SDW}\rangle_{yz}, \quad (5)$$

in which $0 < g, g_1, g_J < 1$ are Gutzwiller factors introduced to describe the correlation effect caused by intra- and inter-orbital Hubbard repulsion and Hund's rule coupling. The operators \hat{D} , \hat{V} and \hat{S} are given by $\hat{D} = \sum_i (n_{i, xz, \uparrow} n_{i, xz, \downarrow} + n_{i, yz, \uparrow} n_{i, yz, \downarrow})$, $\hat{V} = \sum_i (n_{i, xz, \uparrow} + n_{i, xz, \downarrow})(n_{i, yz, \uparrow} + n_{i, yz, \downarrow})$ and $\hat{S} = \sum_i (\vec{S}_{i, xz} \cdot \vec{S}_{i, yz} + \frac{3}{4})$. In the antiferromagnetic ordered ground state, we expect the correction due to the correlation effect to be small as the main effect of the inter-band correlation is embodied

in the mutual enhancement of the AF order in the two bands.

At finite doping, the antiferromagnetic exchange within each band will induce pairing instability in the d-wave channel as in the high- T_c problem. Unlike the high- T_c problem, now we have two degenerate bands which are correlated with each other by the inter-orbital Hubbard repulsion and the Hund's rule coupling. As in the antiferromagnetic ordered state, it is likely that the inter-orbital coupling will induce mutual enhancement of the pairing instability in both bands. However, the inter-orbital coupling will inevitably increase the effective mass of the Cooper pair in each band and thus reduce the superfluid density. With this understanding in mind, we can write down a correlated variational wave function for the superconducting state as we have done for the antiferromagnetic ordered state.

$$|\Psi\rangle = g^{\hat{D}} g_1^{\hat{V}} g_J^{\hat{S}} |d - \text{BCS}\rangle_{xz} \otimes |d - \text{BCS}\rangle_{yz}, \quad (6)$$

in which $|d - \text{BCS}\rangle_{xz}$ and $|d - \text{BCS}\rangle_{yz}$ denote the d-wave BCS superconducting state in the $3d_{xz}$ and the $3d_{yz}$ band. We note that the Gutzwiller factor describing the Hund's rule coupling, $g_J^{\hat{S}}$, now plays a very similar role as that of the projection operator in the construction of the matrix product type ground state of the AKLT model[14]. Thus the superconducting state can be more appropriately called a condensate of entangled Cooper pairs rather than independent Cooper pairs.

The arguments presented above are rather crude and a more careful study of the model proposed in this paper is obviously needed to elucidate its relationship with the superconductivity observed in F-doped LnOFeAs . In principle, the arguments can be made more quantitative through systematic mean field treatment or Gutzwiller approximation. A direct numerical calculation on the variational wave function is also possible. However, before any such effort is made, it is important first to know the actual value of the crystal field splitting by, for example, the optical measurement, and check the validity of neglecting the $3d_{3z^2-r^2}$, $3d_{x^2-y^2}$ and the $3d_{xy}$ orbital in the low energy subspace.

In summary, a microscopic model for the FeAs plane of the newly discovered Iron-based superconductors is proposed. In our model, the orbital degeneracy between the Fe $3d_{xz}$ and the $3d_{yz}$ orbital plays an important role and may explain the peculiarities of the parent compounds of this family of new superconductors. The model pre-

dicts a commensurate antiferromagnetic ordered ground state for the parent compound and a d-wave pairing superconducting ground state in the doped case. In the latter case, the inter-orbital coupling transform the system from a condensate of independent Cooper pairs into a condensate of entangled Coopers and it would be interesting to see effect of inter-orbital couplings on the dynamical properties of the new superconductor.

This work is supported by NSFC Grant No. 10774187. The author acknowledge the discussion with Prof. Zheng-Yu Weng, Prof. Ya-Yu Wang, Prof. Yue-Hua Su and Kai Wu.

-
- [1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *J. Am. Chem. Soc.* 130, 3296 (2008).
 - [2] G. F. Chen, Z. Li, G. Li, J. Zhou, D. Wu, J. Dong, W. Z. Hu, P. Zheng, Z. J. Chen, J. L. Luo and N. L. Wang, *Cond-mat/0803.0128*(2008).
 - [3] Hai-Hu Wen, Gang Mu, Lei Fang, Huan Yang and Xiyu Zhu, *Europhys. Lett.* 82, 17009 (2008); *Cond-mat/0803.3021*(2008).
 - [4] X. H. Chen, T. Wu, G. Wu, R. H. Liu, H. Chen and D. F. Fang, *Cond-mat/0803.3603*(2008).
 - [5] Zhi-An Ren, Jie Yang, Wei Lu, Wei Yi, Xiao-Li Shen, Zheng-Cai Li, Guang-Can Che, Xiao-Li Dong, Li-Ling Sun, Fang Zhou and Zhong-Xian Zhao, *Cond-mat/0803.4283* (2008).
 - [6] D. J. Singh and M.-H. Du, *Cond-mat/0803.0429*(2008).
 - [7] K. Haule, J. H. Shim, and G. Kotliar, *Cond-mat/0803.1279* (2008).
 - [8] Gang Xu, Wenmei Ming, Yugui Yao, Xi Dai, Shou-Cheng Zhang, and Zhong Fang, *Cond-mat/0803.1282* (2008).
 - [9] Chao Cao, P. J. Hirschfeld, and Hai-Ping Cheng, *Cond-mat/0803.3236* (2008).
 - [10] Fengjie Ma and Zhong-Yi Lu, *Cond-mat/0803.3286* (2008).
 - [11] Kazuhiko Kuroki, Seiichiro Onari, Ryotaro Arita, Hidetomo Usui, Yukio Tanaka, Hiroshi Kontani, and Hideo Aoki, *Cond-mat/0803.3325* (2008).
 - [12] Xi Dai, Zhong Fang, Yi Zhou and Fu-chun Zhang, *Cond-mat/0803.3982* (2008).
 - [13] Q. Han, Y. Chen, and Z. D. Wang, *Cond-mat/0803.4346* (2008).
 - [14] I. K. Affleck, T. Kennedy, E. H. Lieb and H. Tasaki, *Commun. Math. Phys.* 115, 477 (1988).